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(54) **Synthetic resin-based dental material composition with antiseptic properties.**

(57) Dental material composition with antiseptic properties, for use in dental restoration and for the production of false teeth, bridges, dental prostheses and the like which comprises, in a synthetic resin base, a quaternary ammonium compound having germicidal and/or bacteriostatic activity, preferably benzalkonium chloride, and a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil.

The composition may also comprise one or more inorganic fillers, and a number of further additives chosen from those known in the art of dental materials, and may be obtained from starting formulations of the two-components type, intended for polymerisation with free-radical initiators, or of the single-component type, intended for polymerisation with photo-initiators.

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The present invention relates to a synthetic resin-based dental material composition with antiseptic properties. More particularly, the invention concerns synthetic polymer materials for use in dental restoration and for the production of false teeth, bridges and dental prostheses, which materials are endowed with inherent germicidal and cariostatic properties.

5 As it is known, tooth crowns, bridges and partial or complete dentures used to be made in the past either of metallic materials such as gold, silver and suitable alloys thereof with other metals, or of ceramic materials such as feldspar or quartz. In order to exploit at one time the good mechanical properties of the metal and the superior appearance of the ceramic material, which can closely reproduce the colour and brightness of natural enamel, metal-ceramic combinations are presently used. They consist of a metallic  
10 structure coated with a thin layer of a suitable ceramic material. However, these combined materials have the drawback of chipping easily, owing to the low tensile strength of the outer ceramic layer.

More recently, synthetic resins have been developed for use as dental materials, and they are now employed not only for restorative fillings and for the restoration of partially damaged teeth, but also for the production of artificial denture bases, of dental crowns, of fixed and removable bridges and dentures. The  
15 synthetic materials most widely used in this field are the acrylic resins, more specifically the resins based on poly(methyl methacrylate), which show a good compatibility with human tissues, good mechanical properties, absence of particular taste or odour once polymerised, low water absorption, good shape stability as well as ability to exactly reproduce shape details, colour stability and acceptable aesthetic appearance, ease of repairing and cleaning.

In order to reduce the shrinkage which occurs during polymerisation of methyl methacrylate, thus reducing at the same time the heat release, the most employed production technique consists in admixing about three parts of acrylic polymer with one part of the methyl methacrylate monomer, so as to obtain a formable dough. The complete polymerisation of the latter is achieved by means of free-radical initiators, such as organic peroxides (e.g., benzoyl peroxide), or by means of photo-initiators, such as some aromatic  
25 ketones (e.g., camphorquinone), in co-operation with UV or visible light.

When polymerisation is started by free-radical initiators the dental material consists of two separate components, one of which contains the polymerisable material while the other contains the catalyst. For heat-cured polymers only a free-radical initiator such as benzoyl peroxide is present as the catalyst component, while for cold-cured polymers an additional ingredient, known as the "activator", is present in  
30 the polymerisable mixture. The activator, usually a tertiary amine such as N,N-dimethyl-p-toluidine, forms a redox system with the free-radical initiator contained in the catalyst component, which redox system starts polymerisation at room temperature.

When polymerisation is started by the action of light the polymerisable dental material is a single-component mixture containing one or more photo-initiators together with the polymerisable ingredients.

35 The synthetic resin-based mixtures for use as dental materials may also contain additional ingredients such as, e.g., cross-linking agents (e.g. ethylene glycol dimethacrylate) and finely divided inorganic material (i.e. the inert fillers), intended to provide the desired colour and mechanical properties, such as hardness, wear resistance and resistance to abrasion.

Besides the acrylic resins, some polycarbonates, resins based on poly(vinyl chloride) (specifically, copolymers of vinyl chloride, vinyl acetate and methyl methacrylate), some polyamides and some  
40 polyurethanes have been tested for their performance as denture base materials and the like, but the results reported have been, so far, not entirely satisfactory.

As concerns the specific field of restorative fillings, the use of poly(methyl methacrylate) resins has been discontinued in favour of specially designed materials whose main monomer component is the so-called Bowen monomer or bis-GMA (or BISGMA, bisphenol glycidyl methacrylate), i.e. the diester of  
45 bisphenol A di(2,3-epoxy propyl)ether with methacrylic acid. Normally, mixtures of BISGMA with diluent monomers such as triethylene glycol dimethacrylate (also known as TEGDMA) are used.

Also in this case, the material comprises inorganic fillers and may contain a number of further additives. The polymerisation may either be started by a free-radical initiator at room temperature (cold-cured  
50 systems, also known as paste-paste systems) or by the action of UV or visible light (light-cured systems, also known as single component systems).

One of the most critical problems encountered in the use of artificial dental elements is that of maintaining a proper oral hygiene. When artificial dental crowns and bridges are present, the gums tend to retract, thus uncovering the gingival groove and the tooth neck, where the bacterial flora more easily finds  
55 the optimal conditions for its development. This is also due to the frequent presence in the said zone of food residues, which escape the usual cleaning. As is known, the foregoing on one hand co-operates in giving rise to problems of unpleasant breath and, on the other hand, may result in paradental inflammations, such as, specifically, gingivitis.

Further, bacterial plaque on natural teeth normally favours the formation of dental cares. The organic acids which are produced by the action of the oral bacteria decalcify the hard tissues of the teeth, thus giving rise to the formation of small cavities wherein the micro-organisms find conditions favourable to their growth. Although dentistry is nowadays provided with means for stopping the caries' progress, by  
 5 destroying the affected tissue starting from the exterior surface towards the interior of the tooth, no effective and readily applicable means have been found up to now for exerting a preventive action against caries, substantially owing to the fact that the knowledge on the etiopathogenesis of this affection is still incomplete.

Besides representing a danger for natural, still undamaged teeth, dental cares may also challenge teeth  
 10 that have already been treated and restored by means of suitable fillings. In spite of the fact that the restoration of a tooth partially damaged by caries is carried out with the utmost care, it may well occur that cares newly develops under the restorative filling, and that the latter is finally lost as a result of the caries' progress.

Therefore, it is an object of the present invention to provide dental material compositions suitable for  
 15 the production of dental prostheses, false teeth, bridges and the like, as well as for use as restorative fillings, which show, in addition to the necessary mechanical and aesthetic features mentioned above, the further property of having a long-lasting antiseptic and germicidal action, so as to hinder the growth of bacterial plaque and prevent the caries formation.

In accordance with the present invention there is suggested to incorporate, in a resin-based formulation  
 20 of the kind suitable for the production of dental elements or fillings, a mixture of essential oils having germicidal and bacteriostatic activity, as well as a quaternary ammonium compound, also with the function of antiseptic.

It is to be noted that a dental element having inherent germicidal activity could not be obtained, e.g., by employing the conventional ceramic materials, since the latter are subjected during production to tempera-  
 25 tures so high that the bactericidal organic compounds mentioned above would surely decompose, thus losing any biological activity.

Therefore, the present invention specifically provides a synthetic resin-based dental material composi-  
 tion characterised in that it also comprises a quaternary ammonium compound having germicidal and/or bacteriostatic activity and a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil.

Preferably, said synthetic resin is chosen from the group consisting of acrylic polymers or copolymers  
 30 (e.g. poly(methyl methacrylate)-based polymers), unsaturated polyester resins, vinyl ester resins, and polymers or copolymers of BISGMA (e.g. copolymers of BISGMA and TEGDMA). Advantageously, said dental material may comprise one or more inorganic fillers chosen from the group consisting of glass micro-  
 beads, quartz particles, corundum particles, micronized silica, fumed silica.

In the present disclosure there is meant by "dental material" any material suitable for the production of  
 35 partial or complete dentures, fixed or removable dental bridges, crowns and the like, as well as any material suitable for being used in dentistry as binding or sealant resin, and for carrying out partial or total restorations of either natural or artificial teeth, such as materials for restorative fillings. Any such element will be also referred to in the following as "dental element".

The quaternary ammonium compound which according to the invention is added to the dental material  
 40 formulation is, advantageously, a quaternary ammonium salt wherein at least one of the four groups bound to the nitrogen atom is a long-chain alkyl group. As a matter of fact, it is known that this class of compounds comprises several products endowed with antibacterial, antifungal and generally disinfecting activity which show an extremely reduced toxicity towards higher organisms. Particularly common examples  
 45 of such compounds are dialkyl-dimethyl ammonium salts (such as, e.g., didecyl-dimethyl ammonium chloride), cetylpyridinium chloride and alkyl-dimethyl-benzyl ammonium salts, including, first of all, benzal-  
 konium chloride. More precisely, the latter is a mixture of alkyl-dimethyl-benzyl ammonium chlorides whose alkyl chains comprise between 8 and 18 carbon atoms. Benzalkonium chloride is known for having a high germicidal activity, even at low concentrations, towards a wide range of micro-organisms, including Gram-  
 50 positive and Gram-negative bacteria.

In the formulation according to the instant invention the disinfecting action of the quaternary ammonium  
 compound is synergically supported by the essential oils. Although some of said essential oils have been known to possess disinfecting and antimicrobial activity since ancient times, the fact that said activity is  
 55 effectively and lastingly exerted when the product is incorporated in a solid and coherent mixture such as  
 that making out a dental element, without negatively affecting, on the other hand, the mechanical properties of said dental element, does not appear to have been disclosed before, and is a specific aspect of this invention.

As it is known, essential oil or essences are active ingredients of vegetable origin which are contained in a great number of plants, from which they can be obtained by pressing or by distillation with water steam or by solvent extraction. The products obtained by extractive means such as solvents or liquid or semiliquid fats are more properly called extracts.

Among the essential oils, mint oil is obtained by steam distillation of the leaves and the flowering tips of the *Mentha piperita* plant, and is a clear colourless or yellowish or some times greenish liquid, whose typical burning odour is followed by a freshness feeling. The essence mainly contains (-)-menthol as the active ingredient, and is used for flavouring toothpastes, confectioneries and chewing gums.

The eucalyptus oils are obtained from the leaves of various plants belonging to the genus *Eucalyptus*, among which, firstly, *Eucalyptus globulus*. The extract of the latter, having a characteristic fresh odour and a colourless or pale yellow aspect, is known as eucalyptole, and contains cineole and  $\alpha$ -pinene. Said product is employed in pharmaceutical preparations as an antiseptic for the respiratory tract.

The bergamot oil is obtained by pressing peels from the *Citrus aurantium* fruit, and is a brownish yellow or greenish yellow liquid, which sometimes contains a deposit. The odour is pleasant, fruity, fresh and sweet. The most important components of bergamot oil are linalyl acetate, linalol and citral, besides the terpene compounds. In addition to being used in perfume production and in cosmetic industry, bergamot oil is also employed in medicinal products, owing to its topical antiseptic properties.

In addition to the final (i.e., already polymerised) dental material compositions mentioned above, this invention also concerns the formulations starting from which said dental material is obtained, i.e., the compositions before the polymerisation step. Specifically, the invention provides compositions for the production of synthetic resin-based dental elements, which compositions may either be of the two-components type, or of the single-component type.

In the former case, as usual, the first component comprises a polymerisable synthetic product and the second component comprises a free-radical polymerisation initiator, and the composition is characterised in that the first component also comprises a quaternary ammonium compound having germicidal and/or bacteriostatic activity and a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil.

In accordance with what set forth before as concerns the preferred synthetic resins for use in the proposed formulations, said polymerisable synthetic product is preferably chosen from the group consisting of acrylic and/or methacrylic compounds, mixtures of unsaturated and saturated carboxylic acids with diols (which give rise by polymerisation to unsaturated polyester resins), mixtures of liquid epoxy resins with acrylic acid (which give rise by polymerisation to vinyl ester resins), BISGMA (i.e. bisphenol glycidyl methacrylate) and mixtures thereof with other dimethacrylates.

The inorganic fillers referred to before may be contained either in said first component or in said second component or in both, as it will be shown in the following examples of two-components formulations according to the invention. All percentage amounts given below are by weight.

#### EXAMPLE 1

Two-components formulation to give poly(methyl methacrylate)-based polymer

• Component 1		
mixture of poly(methyl methacrylate) and methyl methacrylate		qs. to 100%
activator (N,N-dimethyl-p-toluidine)		0.7-1%
mixture of essential oils (mint oil, eucalyptus oil and bergamot oil)		1.2-1.5%
benzalkonium chloride		2.4-3.0%

• Component 2	
benzoyl peroxide	1.7-2.0%
micronized silica	0.9-1.2%
glass micro-beads	qs. to 100%

## EXAMPLE 2

Two-components formulation to give poly(methyl methacrylate)-based polymer

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• Component 1	
methyl methacrylate containing 6% of cross-linking agent (ethylene glycol dimethacrylate)	97-97.9%
activator (N,N-dimethyl-p-toluidine)	0.7-1%
mixture of essential oils (mint oil, eucalyptus oil and bergamot oil)	1.2-1.5%
benzalkonium chloride	0.2-0.5%

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• Component 2	
poly(methyl methacrylate)	qs. to 100%
benzoyl peroxide	0.3-3.1%
quartz or corundum particles	8-10%
glass micro-beads	8-10%
inorganic pigments for various shades	0.8-1.5%

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The above formulation is intended for use in the production of temporary dental prostheses and bridges. About 50% of the whole mass of inert fillers has particle size comprised between 13 and 20  $\mu\text{m}$ . As is usual in the production of dental materials, the inert fillers are previously treated with about 0.025-2% of methacryloxypropyltrimethoxysilane in order to enhance the matrix-to-filler bonding.

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## EXAMPLE 3

Two-components formulation to give unsaturated polyester resins or vinyl ester resins

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• Component 1	
mixture of unsaturated and saturated carboxylic acids with diols, or mixture of liquid epoxy resins with acrylic acid	qs. to 100%
cross-linking agent (styrene)	2.4-3.0%
activator (cobalt octoate)	0.1-0.5%
deaerating agent	0.12-0.15%
mixture of essential oils (mint oil, eucalyptus oil and bergamot oil)	0.2-0.3%
benzalkonium chloride	0.15-0.22%
glass micro-beads	29-35%
micronized silica	0.7-0.9%

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• Component 2	
methyl ethyl ketone peroxide, amount referred to component 1	2-3%

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## EXAMPLE 4

Two-components formulation with BISGMA and TEGDMA (paste-paste system for dental restorations)

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• Component 1	
BISGMA (75%) and TEGDMA (25%)	qs. to 100%
stabiliser (hydroquinone)	0.1%
activator (N,N-dimethyl-p-toluidine)	0.7-1%
mixture of essential oils (mint oil, eucalyptus oil and bergamot oil)	1.2-1.5%
benzalkonium chloride	0.2-0.5%
quartz or corundum particles	30%
glass micro-beads	28%
fumed silica	10%

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• Component 2	
BISGMA (75%) and TEGDMA (25%)	qs. to 100%
benzoyl peroxide	0.3-3.1%
quartz or corundum particles	30%
glass micro-beads	28%
fumed silica	11.7%

Also in this case the inert fillers are previously treated with about 0.025-2% of methacryloxypropyl-trimethoxysilane in order to enhance the matrix-to-filler bonding.

The compositions of the single component type according to the invention comprise, as it is customary, a polymerisable synthetic product and a photo-initiator system, and are characterised in that said single component also comprises a quaternary ammonium compound having germicidal and/or bacteriostatic activity and a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil.

Preferably, said polymerisable synthetic product is chosen from the group consisting of acrylic and/or methacrylic compounds, BISGMA (i.e. bisphenol glycidyl methacrylate) and mixtures thereof with other dimethacrylates.

Light-cured dental compositions intended for being polymerised by the action of UV light generally contain, as the initiator, an aromatic ketone (such as 1,2-diphenyl-2,2-dimethoxy ethanone and 2-methoxy-2-phenyl acetophenone, also known as benzoin-methyl ether), while compositions which may be cured by the action of visible light generally contain a photo-initiator system with two ingredients, generally camphoroquinone and an aliphatic or aromatic amine (such as a N,N-aminoethyl methacrylate), the latter serving as a polymerisation promoter.

Examples of single-component composition according to the invention, for use as restorative filling, are as follows. All percentage amounts given below are by weight.

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## EXAMPLE 5

Single-component formulation with BISGMA and TEGDMA (UV light-cured composite for dental restorations)

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	• Composition	
	BISGMA (75%) and TEGDMA (25%)	qs. to 100%
10	stabiliser (benzophenone)	0.1%
	benzoin-methyl ether	0,2%
	mixture of essential oils (mint oil, eucalyptus oil and bergamot oil)	1.2-1.5%
	benzalkonium chloride	0.2-0.5%
15	quartz or corundum particles	30%
	glass micro-beads	28%
	fumed silica	10%

## EXAMPLE 6

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Single-component formulation with BISGMA and TEGDMA (visible light-cured composite for dental restorations)

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	• Composition	
	BISGMA (75%) and TEGDMA (25%)	qs. to 100%
	stabiliser (benzophenone)	0.1%
30	camphoroquinone	0,2%
	N,N-aminoethyl methacrylate	0,1%
	mixture of essential oils (mint oil, eucalyptus oil and bergamot oil)	1.2-1.5%
	benzalkonium chloride	0.2-0.5%
	quartz or corundum particles	30%
35	glass micro-beads	28%
	fumed silica	10%

As pointed out, the formulations disclosed in examples 5 and 6 are suitable for use, respectively, with UV light sources and with sources irradiating visible light. It is also possible, in order to avoid the need of differentiated restorative filling products depending on the light source available to the dentist, to provide a product according to the invention which includes in one only formulation both initiator systems.

The production techniques of dental elements by employing the formulations according to this invention are not substantially different from those known in the art. The only specific aspect that should be taken into account is the limited resistance of the organic antiseptic compounds of the invention to severe temperature conditions. For instance, in the production of acrylic resin temporary prostheses the exothermic polymerisation reaction brings about an increase in the reaction mixture temperature, which rapidly reaches a peak level of 38-40 °C, and then slowly returns to room levels. It is important, as pointed out before, that the exothermic peak does not exceed the said values.

By way of an example, the results of some experimentations carried out on samples of antiseptic dental material according to the invention are set forth below. The active materials subjected to the tests have been obtained from the polymerisation of two components formulated as follows.

	• Component 1	
55	mixture of poly(methyl methacrylate) and methyl methacrylate	250 g
	mixture of essential oils (mint oil, eucalyptus oil and bergamot oil)	3.54 g
	benzalkonium chloride	7.10 g

• Component 2	
glass micro-beads (62-200 $\mu\text{m}$ )	375 g
benzoyl peroxide	7.50 g
micronized silica	4.16 g

Said material has been used to produce round plates 4 cm in diameter and some millimetres in thickness. The control plates, of the same shape and size, only contain the resin and the inert fillers. The features of four of said sample plates are as follows:

- sample No. 1 control
- sample No. 2 control
- sample No. 3 composition according to the invention produced some days before the test
- sample No. 4 composition according to the invention produced more than 1 year before the test

In order to evaluate the action of the above samples on micro-organisms, bacterial strains have been used which had been isolated from clinical materials (more precisely, oropharyngeal exudates) of out-patients.

The bacterial species examined are the following:

- alpha-haemolytic streptococci;
- Staphylococcus epidermidis*;
- beta-haemolytic streptococci;
- Staphylococcus aureus*;
- Pseudomonas aeruginosa*;
- Klebsiella pneumoniae*;
- Proteus mirabilis*.

All of the strains have been isolated on solid media and have been identified through their morphological and cultural characteristics, and by means of biochemical tests. From all of the strains suspensions in sterile distilled water have been prepared, so as to obtain a turbidity of  $7.5 \times 10^6$  cells/ml as measured on the McFarland turbidimetric scale. After dividing each plate into 7 circular sectors, each one of the bacterial suspensions prepared has been placed in one sector of each plate, by means of a 10  $\mu\text{l}$  graduated pipette. The samples so treated have been left for 4 hours closed in sterile Petri dishes, on the bottom of which some blotting paper impregnated with sterile distilled water had been placed in order to moisturise the interior of the dishes. The temperature was the room temperature, oscillating between 18 °C and 20 °C.

After 4 hours some sterile swabs were passed over each sector of the plates, which swabs were dissolved in 22 ml of tripton soya broth. The resulting mixture was incubated at 37 °C for 24 hours. Subsequently, the broth cultures were used for seeding on selective means, in order to identify the micro-organisms. The culture means employed were the following:

- alpha-haemolytic streptococci: modified CNA Columbia agar-blood;
- Staphylococcus epidermidis*: mannite salt agar;
- beta-haemolytic streptococci: modified CNA Columbia agar-blood;
- Staphylococcus aureus*: mannite salt agar;
- Pseudomonas aeruginosa*: cetrimide agar;
- Klebsiella pneumoniae*: McConkey agar;
- Proteus mirabilis*: McConkey agar.

The results obtained from the above experimental procedure are set forth in the following table, wherein "-" represents no micro-organisms growth, "+" represents a somewhat limited growth, "++" and "+++" respectively represent a good growth and a particularly abundant growth.



TABLE 1

Micro-organisms growth on acrylic resin-based plates				
MICRO-ORGANISM	SAMPLE No.			
	1	2	3	4
alpha-haemolytic streptococci	+	+	-	-
Staphylococcus epidermidis	+	+	-	-
beta-haemolytic streptococci	+	-	-	-
Staphylococcus aureus	+	-	-	-
Pseudomonas aeruginosa	+	+	-	-
Klebsiella pneumoniae	+	+	-	-
Proteus mirabilis	+	+	-	-

The results reported above confirm the germicidal activity of the resin-based compositions according to the invention. In fact, both in sample 3 and in sample 4 benzalkonium chloride and the essential oils have been shown to greatly interfere with the growth of the bacterial strains placed on the sample itself: none of the seedings carried out on each sector of the examined plates appears to have resulted in the growth any micro-organism, either Gram-positive or Gram-negative.

On the contrary, on all of the sectors of the control plate No. 1 an abundant bacterial growth took place. As far as the control plate No. 2 is concerned, the growth of the three Gram-negative species (i.e., *Ps. aeruginosa*, *K. pneumoniae* and *P. mirabilis*), of a quite low number of colonies of alpha-haemolytic streptococci and of *Staphylococcus epidermidis* was detected, while beta-haemolytic streptococci and *Staphylococcus aureus* did not develop at all. It is to be noted that before the test was carried out the sample No. 2 had been left for some time into contact with one of the samples of the material according to the invention, since the plates had been kept stacked one over the other; the fact that the same microbial strains, in the same conditions, abundantly grew on plate No. 1 but not as much on plate No. 2 shows that the simple contact with the proposed germicidal material may negatively affect the bacterial growth even on substrates which have not the composition according to the invention. This allows to foresee that the presence in the oral cavity of a dental element according to the invention may exert a preventive anti-caries action towards the natural teeth located close to said dental element.

Coming back to samples 3 and 4, made of the proposed dental material, it is worthwhile noting that the sample No. 4, which had been produced more than one year before the test, still showed the same activity level as the freshly made sample. The foregoing is an evidence of the fact that the antiseptic activity of the materials according to the invention lasts for a long time. Such property is clearly fundamental in view of the average life time required to dental elements such as bridges, crowns, prostheses and restorative fillings.

#### Claims

1. Synthetic resin-based dental material composition, characterised in that it also comprises a quaternary ammonium compound having germicidal and/or bacteriostatic activity and a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil.
2. Composition according to claim 1 wherein said synthetic resin is chosen from the group consisting of: acrylic polymers or copolymers, unsaturated polyester resins, vinyl ester resins, polymers or copolymers of BISGMA (i.e. bisphenol glycidyl methacrylate).
3. Composition according to claim 2 wherein said acrylic polymer or copolymer is a poly(methyl methacrylate)-based polymer.
4. Composition according to claim 2 wherein said polymer or copolymer of BISGMA is a copolymer of BISGMA and TEGDMA (i.e. triethylene glycol dimethacrylate).
5. Composition according to any one of claims 1-4, wherein said quaternary ammonium compound is benzalkonium chloride.

6. Composition according to any one of claims 1-5, also comprising one or more inorganic fillers chosen from the group consisting of: glass micro-beads, quartz particles, corundum particles, micronized silica, fumed silica.
- 5 7. Composition for the production of synthetic resin-based dental elements, said composition being of the two-components type, wherein the first component comprises a polymerisable synthetic product and the second component comprises a free-radical polymerisation initiator, characterised in that said first component also comprises a quaternary ammonium compound having germicidal and/or bacteriostatic activity and a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil.
- 10 8. Composition according to claim 7, wherein said polymerisable synthetic product is chosen from the group consisting of: acrylic and/or methacrylic compounds, mixtures of unsaturated and saturated carboxylic acids with diols, mixtures of liquid epoxy resins with acrylic acid, BISGMA (i.e. bisphenol glycidyl methacrylate) and mixtures thereof with other dimethacrylates.
- 15 9. Composition according to any one of claims 7-8 wherein said quaternary ammonium compound is benzalkonium chloride.
- 20 10. Composition according to any one of claims 7-9 wherein either said first component or said second component or both also comprise one or more inorganic fillers chosen from the group consisting of: glass micro-beads, quartz particles, corundum particles, micronized silica, fumed silica.
- 25 11. Composition according to claim 10 wherein said first component comprises a mixture of poly(methyl methacrylate) and methyl methacrylate, benzalkonium chloride, a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil and a polymerisation activator, and said second component comprises benzoyl peroxide, glass micro-beads and micronized silica.
- 30 12. Composition according to claim 10 wherein said first component comprises methyl methacrylate, benzalkonium chloride, a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil, a cross-linking agent and a polymerisation activator, and said second component comprises poly(methyl methacrylate), benzoyl peroxide, glass micro-beads and quartz or corundum particles.
- 35 13. Composition according to claim 10 wherein said first component comprises a mixture of unsaturated and saturated carboxylic acids with diols, or a mixture of liquid epoxy resins with acrylic acid, benzalkonium chloride, a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil, glass micro-beads, micronized silica, a cross-linking agent and a polymerisation activator, and said second component comprises methyl ethyl ketone peroxide.
- 40 14. Composition according to claim 10 wherein said first component comprises a mixture BISGMA (i.e. bisphenol glycidyl methacrylate) and TEGDMA (i.e. triethylene glycol dimethacrylate), benzalkonium chloride, a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil, quartz or corundum particles, glass micro-beads, fumed silica and a polymerisation activator, and said second component comprises a mixture of BISGMA (i.e. bisphenol glycidyl methacrylate) and TEGDMA (i.e. triethylene glycol dimethacrylate), benzoyl peroxide, quartz or corundum particles, glass micro-beads, and fumed silica.
- 45 15. Composition for the production of synthetic resin-based dental elements, said composition being of the single component type, wherein said single component comprises a polymerisable synthetic product and a photo-initiator system, characterised in that said single component also comprises a quaternary ammonium compound having germicidal and/or bacteriostatic activity and a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil.
- 50 16. Composition according to claim 15, wherein said polymerisable synthetic product is chosen from the group consisting of: acrylic and/or methacrylic compounds, BISGMA (i.e. bisphenol glycidyl methacrylate) and mixtures thereof with other dimethacrylates.
- 55 17. Composition according to any one of claims 15-16 wherein said quaternary ammonium compound is benzalkonium chloride.

18. Composition according to any one of claims 15-17 wherein said single component also comprises one or more inorganic fillers chosen from the group consisting of: glass micro-beads, quartz particles, corundum particles, micronized silica, fumed silica.

5 19. Composition according to claim 18 wherein said single component comprises a mixture of BISGMA (i.e. bisphenol glycidyl methacrylate) and TEGDMA (i.e. triethylene glycol dimethacrylate), benzalkonium chloride, a mixture of essential oils comprising mint oil, eucalyptus oil and bergamot oil, quartz or corundum particles, glass micro-beads, fumed silica and a photo-initiator system.

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European Patent  
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## EUROPEAN SEARCH REPORT

Application Number  
EP 94 83 0479

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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
Y	GB-A-2 193 967 (G C DENTAL IND CORP) 24 February 1988 * page 2, line 61 - page 3, line 9 * * page 5, line 19 - line 33 * * claims * ---	1-19	A61K6/083
Y	WO-A-93 08792 (WARNER LAMBERT CO) 13 May 1993 * page 4, line 12 - line 23 * * page 8, line 26 - page 9, line 8 * * page 11, line 1 - line 3 * ---	1-19	
Y,P	WO-A-94 18939 (WARNER LAMBERT CO) 1 September 1994 * claims * ---	1-19	
A	EP-A-0 264 660 (IVOCLAR AG) 27 April 1988 * page 3, line 55 - page 4, line 4 * ---	1	
A	US-A-5 080 583 (HUNTING TANYA L) 14 January 1992 -----		
			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			A61K
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 11 July 1995	Examiner Cousins-Van Steen, G
<b>CATEGORY OF CITED DOCUMENTS</b> X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons ----- & : member of the same patent family, corresponding document			